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09/809,423	03/16/2001	Jens Klein		4925

7590 06/21/2004  
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EXAMINER

SODERQUIST, ARLEN

ART UNIT	PAPER NUMBER
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1743

DATE MAILED: 06/21/2004

Please find below and/or attached an Office communication concerning this application or proceeding.

## Office Action Summary

**Application No.**

09/809,423

**Applicant(s)**

KLEIN ET AL.

**Examiner**

Arlen Soderquist

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-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --  
**Period for Reply**

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If the period for reply specified above is less than thirty (30) days, a reply within the statutory minimum of thirty (30) days will be considered timely.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

**Status**

- 1) ☒ Responsive to communication(s) filed on 08 April 2004.
- 2a) ☐ This action is **FINAL**. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

**Disposition of Claims**

- 4) ☒ Claim(s) 39-76 is/are pending in the application.
- 4a) Of the above claim(s) \_\_\_\_\_ is/are withdrawn from consideration.
- 5) ☐ Claim(s) \_\_\_\_\_ is/are allowed.
- 6) ☒ Claim(s) 39-76 is/are rejected.
- 7) ☐ Claim(s) \_\_\_\_\_ is/are objected to.
- 8) ☐ Claim(s) \_\_\_\_\_ are subject to restriction and/or election requirement.

**Application Papers**

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on \_\_\_\_\_ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.  
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).  
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

**Priority under 35 U.S.C. § 119**

- 12) ☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☒ All b) ☐ Some \* c) ☐ None of:
1. ☒ Certified copies of the priority documents have been received.
  2. ☐ Certified copies of the priority documents have been received in Application No. \_\_\_\_\_.
  3. ☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

\* See the attached detailed Office action for a list of the certified copies not received.

**Attachment(s)**

- |  |   |
|--|---|
| 1) <input checked="" type="checkbox"/> Notice of References Cited (PTO-892)  | 4) <input type="checkbox"/> Interview Summary (PTO-413)<br>Paper No(s)/Mail Date. _____ |
| 2) <input type="checkbox"/> Notice of Draftsperson's Patent Drawing Review (PTO-948)                                   | 5) <input type="checkbox"/> Notice of Informal Patent Application (PTO-152)             |
| 3) <input type="checkbox"/> Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08)<br>Paper No(s)/Mail Date _____ | 6) <input type="checkbox"/> Other: _____  |

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1. A request for continued examination under 37 CFR 1.114, including the fee set forth in 37 CFR 1.17(e), was filed in this application after final rejection. Since this application is eligible for continued examination under 37 CFR 1.114, and the fee set forth in 37 CFR 1.17(e) has been timely paid, the finality of the previous Office action has been withdrawn pursuant to 37 CFR 1.114. Applicant's submission filed on April 8, 2004 has been entered.

2. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries set forth in *Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
  2. Ascertaining the differences between the prior art and the claims at issue.
  3. Resolving the level of ordinary skill in the pertinent art.
  4. Considering objective evidence present in the application indicating obviousness or nonobviousness.
3. Claims 39-76 are rejected under 35 U.S.C. 103(a) as being unpatentable over Willson (either WO 97/32208 (Willson '208) or US 6,063,633 Willson '633)) or Akporiaye (US 6,627,445, newly cited and applied) in view of Fawcett, Clausen, Hunger, Latzel, LeBlond, Moon, Newman, Topsøe or Watanabe (last 8 newly cited and applied).

Both Willson '208 and Willson '633 come from the same application and therefore contain the same disclosure. While this explanation of the Willson references will refer to the Willson '633 patent, corresponding disclosure is found in the Willson '208 application. Willson '633 teaches a catalyst testing process and apparatus. In column 1 lines 27-40, Willson '633 teaches that catalyst testing was conventionally accomplished in bench scale or larger pilot plants in which the feed is contacted with a catalyst under reaction conditions, generally with effluent products being sampled, often with samples being analyzed and results subjected to data

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resolution techniques. Those procedures can take a day or more for a single run on a single catalyst. While those techniques have value in fine-tuning the optimum matrices, pellet shape, etc., the Willson '633 invention permits the scanning of dozens of catalysts in a single set-up, ***often in less time than required for a single catalyst to be evaluated by conventional methods.*** Further, when practiced in its preferred robotic embodiments, the invention can sharply reduce the labor costs per catalyst screened. In the apparatus and method a multicell holder e.g. a honeycomb (microchannel array) or plate, or a collection of individual support particles, is treated with solutions/suspensions of catalyst ingredients to produce cells, spots or pellets holding each of a variety of combinations of the ingredients, is dried, calcined or treated as necessary to stabilize the ingredients in the cells, spots or pellets, then is contacted with a potentially reactive feed stream or batch e.g., biochemical, gas oil, hydrogen plus oxygen, propylene plus oxygen,  $\text{CCl}_2\text{F}_2$  and hydrogen, etc. The reaction occurring in each cell can be measured, e.g. by infrared thermography, spectroscopic detection of products or residual reactants, or by sampling, e.g. by multistreaming through low volume tubing, from the vicinity of each combination, followed by analysis e.g. spectral analysis, chromatography etc, or by observing temperature change in the vicinity of the catalyst e.g. by thermographic techniques, to determine the relative efficacy of the catalysts in each combination. Robotic techniques can be employed in producing the cells, spots, pellets, etc. Columns 2-3 summarize some of the aspects of the invention including reaction types, sensors, catalyst taggants and reactions conditions. The reaction types include any reaction which can be enhanced by the presence of a catalyst such as polymerization reactions, halogenation, oxidation, hydrolysis, esterification, reduction and any other conventional reaction which can benefit from a catalyst. Hydrocarbon conversion reactions, as used in petroleum refining are an important use of the invention and include reforming, fluid catalytic cracking, hydrogenation, hydrocracking, hydrotreating, hydrodesulfurizing, alkylation and gasoline sweetening. The sensors used to detect catalytic activity in the candidate catalysts include chromatographs, temperature sensors, and spectrometers. Especially those adapted to measure temperature and/or products near each specific catalyst spot e.g. by multistreaming, multitasking, sampling, fiber optics, or laser techniques such as thermography, as by an infrared camera recording the temperature at a number of catalyst sites simultaneously, NMR, NIR, TNIR, electrochemical, fluorescence

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detectors, Raman, flame ionization, thermal conductivity, mass, viscosity and stimulated electron or X-ray emission. Optionally taggants (labels) can be added to identify particular catalysts, particularly where particles are employed as supports for the catalysts. These taggants can be conventional as discussed in the literature. Taggants can be chemicals which are stable at reaction conditions or can be radioactive with distinctive emissions. The techniques of combinatorial chemistry will be applicable with taggants as well as with catalysts chosen to suit the particular reaction to be enhanced by the catalyst. Temperatures, pressures, space velocities and other reaction conditions can be varied and will be determined by the reactants and reaction. Willson '633 does not give examples of using two analysis methods together.

In the patent Akporiaye teaches apparatus and method for simultaneously evaluating a plurality of catalysts. The process involves containing the plurality of catalysts in an array of parallel reactors with each reactor containing a bed of catalyst. Each bed of catalyst is then simultaneously contacted, at reaction conditions, with a reactant to form an effluent of each reactor. The reactant or an inert fluid is at a space velocity sufficient to fluidize the catalyst beds. Each of the effluents is analyzed. Column 8, line 40 to column 9, line 47 discusses the analysis of the effluents and teaches that the effluents produced are *analyzed using at least one analytical technique* to determine whether products have been formed, how much product has been formed, and/or which specific product compounds have been formed. *The analytical technique used may be any suitable technique for the type of information desired and components involved.* Preferred techniques include, generally, chromatography, spectroscopy, and nuclear magnetic resonance. Various different forms of chromatography and/or spectroscopy may be employed. Examples include liquid chromatography, gas chromatography, ultraviolet absorption spectroscopy, visible absorption spectroscopy, ultraviolet-visible spectroscopy, atomic absorption spectroscopy, infrared absorption spectroscopy, and emission spectroscopy. While chromatography and spectroscopy methods are preferred, other acceptable techniques include but are not limited to fluorescence spectrometry, mass spectrometry, X-ray methods, radiochemical methods, electroanalytical methods, potentiometric methods, conductometric methods, electrogravimetric methods, coulometric methods, and voltammetry. At least a portion of the effluent from each reactor is conveyed to the analytical instrument. The effluents may be directly conducted to an analytical instrument, or aliquots of the effluents may be sampled and

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delivered to the location of the analytical instrument. The effluents may also be analyzed on stream as they are removed from the reactors. In evaluating catalyst performance, observing trends of activity, selectivity, and yield over time is valuable. Therefore, the effluent being withdrawn from each reactor may be periodically or continuously analyzed. The selectivity, activity, and/or yield may be determined at each analysis time, and the trend of the selectivity and activity may then be observed over time. It is preferred that the effluents of each of the reactors be sampled simultaneously. The benefit of simultaneous sampling is that the results from each catalyst bed are more readily comparable since each catalyst bed would be exposed to the reactant for the same period of time. For quantitative results, the amounts of the effluents analyzed are measured. The specific analysis performed depends upon the application and the desired information. For example, if only the activity of the plurality of catalysts are to be determined and compared, an analysis measuring the amount of reactant consumed in each effluent may be sufficient. Also, a qualitative analysis for the composition of the effluent could be used as an indication of catalyst activity. However, it is generally preferred to have both activity and selectivity information and, in that case, the analytical technique would be selected to measure the quantity of the different components present in each effluent. Using both the activity information and the selectivity information, the yield to the desired products can be calculated and compared between the individual catalysts or mixtures of catalysts that make up the plurality of catalysts. Akporiaye does not give examples of using two analysis methods together.

In the paper Fawcett describes a new instrument. Three powerful analytical techniques, differential scanning calorimetry (DSC), x-ray diffraction (XRD), and mass spectrometry (MS) were combined so that one can completely characterize materials as they are heated in controlled atmospheres. The XRD capability tells, continuously, about the structure of the solid phase in the reaction chamber. The MS monitors the volatiles. And the DSC tells about reaction and phase-change thermochemistry. The set-up was used to study the melting behavior of polymers, the mechanisms of reduction in copper catalysts, and the thermal processing of pharmaceuticals. The combined instrument has several advantages over analyses where the 3 techniques are run separately. The simultaneous analysis allows the analyst to assign specific structural or chemical process data directly to observed thermal events. Because the same environment and sample are

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used for all 3 analyses, instrumental and sample preparation conditions and errors associated with them are kept constant.

In the paper Clausen teaches an in situ cell for combined XRD and on-line catalysis tests of copper-based water gas shift and methanol catalysts. A newly developed in situ x-ray diffraction (XRD) cell was used to obtain information on the structure of binary Cu-Zn and ternary Cu-Zn-Al catalysts during reduction and water gas shift and MeOH synthesis. A major advantage of the cell is that it also serves as an ideal plug flow catalytic reactor such that realistic catalytic and structural information can be obtained simultaneously on the same sample. The cell can be operated both at high temperatures and high pressures. Direct MeOH activity tests confirmed the suitability of the cell. By use of x-rays from a synchrotron source, dynamic studies on the time scale of seconds are demonstrated. This feature was used to study the phase transformation occurring during the activation of the calcined catalysts. In the active catalysts, Cu metal is the only crystalline Cu phase observed, and the formation of this phase is closely related to the disappearance of CuO in the calcined catalyst. The XRD results provide detailed information on the nucleation and growth processes. The variation in the water gas shift activity correlates with the changes in the Cu surface area.

In the paper Hunger presents a technique for simultaneous in-situ MAS NMR and online gas chromatographic studies of hydrocarbon conversions on solid catalysts under flow conditions. A new technique was introduced for simultaneous in-situ MAS NMR investigations of hydrocarbon conversions on solids under flow conditions and online gas chromatography. For adsorption of MeOH on zeolite H-Beta, equal amounts of adsorbed molecules were determined by both analytical methods. Studying the synthesis of Me tert-Bu ether (MTBE) on zeolite H-Beta using an MAS NMR rotor reactor, a constant yield of MTBE of 27% was obtained up to a weight hourly space velocity of  $1.4 \text{ h}^{-1}$ . The variation of the reaction temperature led to a simultaneous change of the  $^{13}\text{C}$  MAS NMR signals of isobutoxy species and of the yield of MTBE determined by online gas chromatography, which indicated that isobutoxy species act as the chemically active compounds. In this first application, the new in-situ technique has demonstrated its advantage for a simultaneous investigation of compounds with a long residence time on the catalyst surface and of compounds rapidly leaving the catalyst surface.

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In the paper Latzel teaches a rapid method for catalyst activity measurements by combination of pulse reactor, mass spectrometer and process computer. The combination of a pulse reactor attached directly (without a separation column) to a mass spectrometer-process computer system is described. In order to obtain a conversion-temperature diagram for the dehydration and dehydrogenation of 2-butanol <1 h is required. Therefore, the method can be used for rapid characterization or comparison of the activities of various samples. The figure of page 394 and its associated discussion teach a temperature measurement at the same time the conversion is being measured by the mass spectrometer.

In the paper LeBlond presents a combined approach to characterization of catalytic reactions using in situ kinetic probes. Several in situ probes for continuously monitoring rate of catalytic reactions under reaction conditions are described. They are reaction calorimetry, measurements of hydrogen uptake in the case of hydrogenation, and IR spectroscopy. In studying catalytic hydrogenation reactions, for example, these in situ probes provide kinetic details of the reactions from different perspectives over the entire course of the reaction. The reaction calorimetry and the hydrogen uptake measure directly, continuously, and in a non-invasive manner the rate of reaction, while the in situ IR spectroscopy provides time-resolved compositional information in the liquid phase. A combination of the information thus obtained leads to a clear and coherent kinetic picture of the reaction under study which can greatly facilitate pathway analysis and mechanistic description of the catalytic reaction. In this report, the usefulness of the combination of these in situ probes is illustrated with two examples of heterogeneously-catalyzed hydrogenation reactions.

In the patent Moon describes a catalyst characterization apparatus capable of characterizing the surface of a catalyst more accurately by a volumetric method without requiring a pre-treatment step which may vary the characteristics of the catalyst as well as without exposing the catalyst in air, by combining a dynamic flow type reactor with a volumetric type adsorption apparatus. With this apparatus it is possible to accurately characterize the catalyst during an actual reaction. It is possible to accurately characterize the various catalysts and to characterize the catalyst during the reaction, alternately and/or continuously, by combining a dynamic flow type reactor with a volumetric type characterization apparatus as well



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as a dynamic flow type characterization apparatus. The figures show both a gas chromatograph (16) and mass spectrometer (17) connected to the reactor (14) in parallel.

In the paper Newman presents a study of model copper-based catalysts by simultaneous differential scanning calorimetry, x-ray diffraction and mass spectrometry. The results of this study show  $\text{Cu}_2\text{O}$  as an intermediate in the reduction of the  $\text{CuO}$  portion of each of the model catalysts. Such features as the onset temperature and Cu surface area varied widely among the 4 catalysts in response to the same chemical event conducted under similar experimental conditions. Oxidation runs on the reduced catalysts were all similar, 1 producing  $\text{Cu}_2\text{O}$  from metal over a broad range of temperature, followed by the oxidation of  $\text{CuO}$  at even higher temperatures.

In the paper Topsøe teaches combined in-situ FTIR and on-line activity studies applied to vanadia-titania  $\text{DeNO}_x$  catalyst. An approach for performing combined in-situ FTIR and online activity measurements is described. The application of such studies to  $\text{V}_2\text{O}_5\text{-TiO}_2$   $\text{DeNO}_x$  catalysts is given. The trends in catalytic activity measured directly with the FTIR cell/reactor agreed with separate activity measurements demonstrating that the present experimental approach provides a direct link between the surface chemistry and the catalysis. During the  $\text{DeNO}_x$  reaction conditions, both Brønsted and Lewis acid sites are present on the surface and they adsorbed  $\text{NH}_3$  strongly. The surface V-OH and V = O groups are probably involved in the catalytic reaction.

In the paper Watanabe discusses dissociation reactions of carbon monoxide gas on iron and iron oxide ( $\text{Fe}_2\text{O}_3$ ) surfaces observed by Raman-ellipsometry spectroscopy. A combined system of Raman spectroscopy and ellipsometry was developed for the study of catalyst surfaces on which a chemical reaction is taking place. The dielectric function and the thickness of a surface layer or a surface compound produced in the reaction are analyzed by ellipsometry; vibrational modes of the compound and surface species are analyzed by Raman spectroscopy, and the products in the gas phase by mass spectrometry. The system was applied to the study of the Boudouard and dissociation reactions of CO molecules in Fe and  $\text{Fe}_3\text{O}_4$  catalysts. A surface layer produced by diffused C and O atoms, a thin oxide layer produced on Fe by CO dissociation, and graphite layers produced by the Boudouard reaction were analyzed. Raman spectra reveal the existence of several kinds of C species, such as defective graphite and an iron

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carbide formed on the Fe and Fe<sub>3</sub>O<sub>4</sub> surfaces. Several Raman bands may be due to microscopic clusters of surface graphite. The reaction of the defective graphite with H<sub>2</sub> gas is stepwise on a time scale of seconds.

It would have been obvious to one of ordinary skill in the art at the time the invention was made to use a combination of appropriate analysis methods taught by any of Willson '633, Willson '203, Akporiaye, Clausen, Hunger, Latzel, LeBlond, Moon, Newman, Topsøe or Watanabe in the Willson '633, Willson '203 or Akporiaye methods and apparatus because of the clear advantages taught by Fawcett, Clausen, Hunger, Latzel, LeBlond, Moon, Newman, Topsøe or Watanabe when more than one analysis method is used on the same sample.

4. Applicant's arguments filed April 8, 2004 have been fully considered but they are not persuasive. The anticipation rejection has been withdrawn because applicant has perfected their claim to the date of the priority document. Relative to the obviousness rejection applicant is reminded that Willson is an advance on single reactions run separately and as such would have lead one to use the possibility of multiple detectors use to gain the advantages taught by Fawcett for multiple detectors. Additionally, the newly cited and applied Akporiaye reference clearly teaches at least one detector in combination with the use of an appropriate detection method to obtain the information desired. The additional newly cited and applied references clearly show that one of skill in the art would have understood the benefit of using a plurality of detectors for simultaneous analysis of the catalyst and or the various aspects of the catalytic reaction. It should also be pointed out the Willson is concerned with or sees that one of the advantages with his method is the processing of a plurality of catalyst samples within the time it took the conventional methods to analyze one catalyst. In this same manner one of skill in the art would have expected a time savings since multiple analyses are conducted concurrently giving complementary information. Relative to the second sensor, it should be pointed out that a different parameter could be the concentration of a product that is not measured by the first sensor. Or it could be that the first sensor is measuring the presence of a product such as a UV spectrometer while the second sensor measures the molecular weight of the product(s) measured by the UV sensor. Thus the scope of the instant language includes the sensor combinations which applicant argues against.

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5. The prior art made of record and not relied upon is considered pertinent to applicant's disclosure. The additionally cited art relates to detectors used in catalyst testing. It is noted that some have multiple detectors as those that have been applied.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Arlen Soderquist whose current telephone number is (571) 272-1265 as a result of the examiner moving to the new USPTO location. The examiner's schedule is variable between the hours of about 5:30 AM to about 5:00 PM on Monday through Thursday and alternate Fridays.

A general phone number for the organization to which this application is assigned is (571) 272-1700. The fax phone number to file official papers for this application or proceeding is (703) 872-9306.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).



June 17, 2004

ARLEN SODERQUIST  
PRIMARY EXAMINER